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Fuel-Neutral Studies of Particulate Matter Transport Emissions

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Project ID # ACS056

PNNL

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This work was funded by the U.S. Department of Energy (DOE) Vehicle Technologies Program

Overview

Timeline

- Start - FY16*
- Finish - FY18

* Three-year scope proposed in response to 2015 National Laboratory Call

Barriers

- Lack of fundamental understanding of factors affecting filter mass and number efficiency
- Need for better models of aftertreatment components

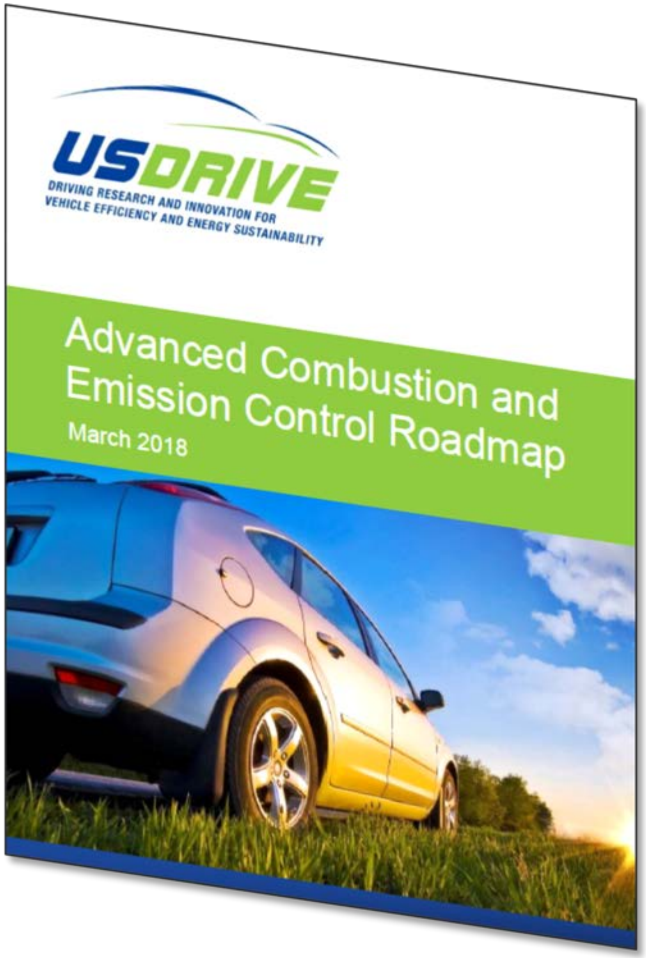
Budget

- Funding received in FY17 - $250K
- Funding received in FY18 - $108K

Partners

- General Motors Company - provide project guidance, support for ERC
- Engine Research Center at University of Wisconsin, Madison - host and operate test engines, perform experiments
- Massachusetts Institute of Technology - Micro X-Ray CT

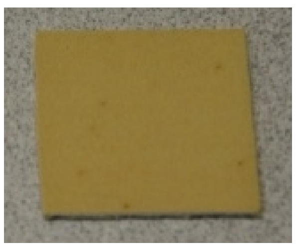
Relevance

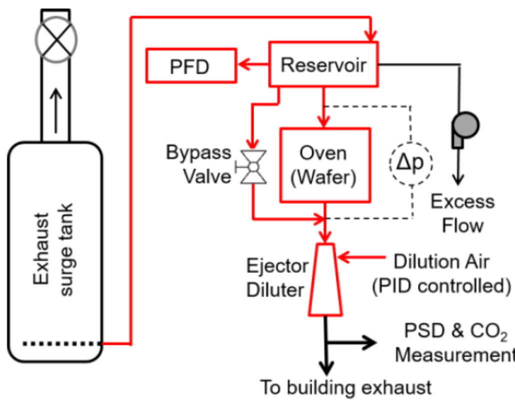


- Barriers:
  - "Worldwide, regulations for particulate matter are both mass- and number-based. Therefore, both the engine-out and filter-out particulate mass and number need to be characterized. In addition, the filtration efficiency on both a mass- and number-basis needs to be understood."
  - "While significant progress has been made in developing models of aftertreatment components and systems, more progress is needed in this important area, as engine/aftertreatment manufacturers have become increasingly reliant on simulation for design and development of products."
- Technical Strategies:
  - "Develop robust simulation tools for individual catalyst aftertreatment and particulate filter components/systems"
  - "Validate models with experimental approaches that further fundamental understanding"

Approach

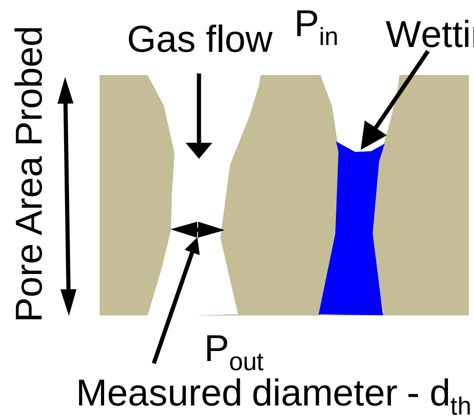
- Exhaust Filtration Analysis (EFA) filtration experiments at U of Wisconsin, Madison
  - Flat wafer filter samples – single wall
  - Particulates from single cyl SIDI test engine
  - Focus on low (but non-zero) soot loadings
  - Completing rebuild of engine and EFA cart

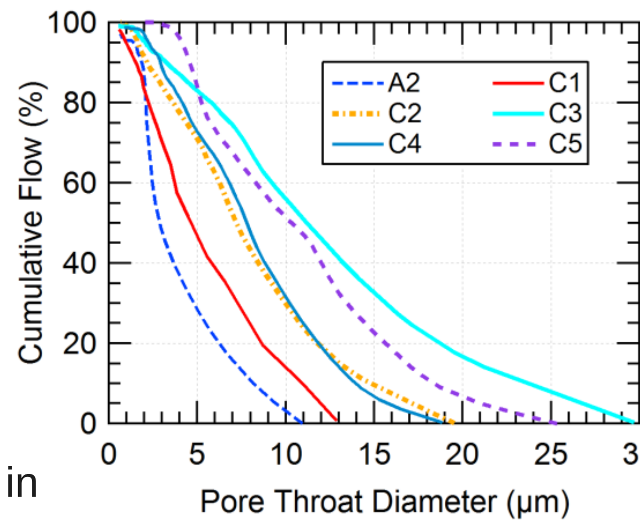




- Filter characterization
  - Micro X-Ray CT
  - Mercury intrusion porosimetry (MIP)
  - Capillary flow porometry (CFP)
    - CFP directly provides information on throat diameter in through-pores needed for novel filtration models
    - CFP provides throat size distributions

Capillary Flow Porometry





Approach

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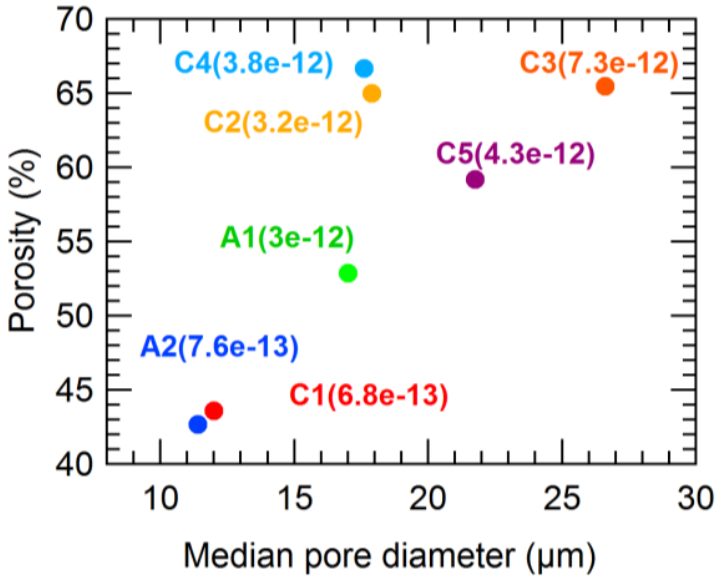
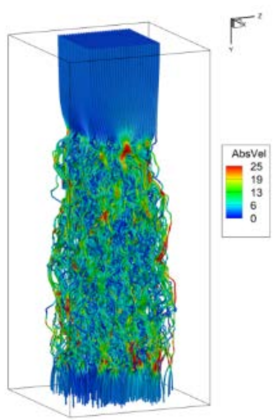
- Micro X-ray computed tomography provides 3D information about pore shape and connectivity
- Backpressure and filter efficiency are determined by 3D flow and transport effects, which can be examined and compared using the lattice-Boltzmann (LB) method
- The ultimate goal is to develop improved filter modeling tools for better system design
- A single model may not be suited for all purposes



Date	Milestone	Status
3/30/18	Develop method to characterize participating pore volume in filter substrates using 3D lattice Boltzmann flow field solutions.	Complete
6/30/18	Complete Eulerian filtration simulations of at least six substrates and compare to experimental data.	Complete
6/30/18	Use 3D micro-scale simulations to predict permeability of catalyzed and ash-coated filters using CT data provided by Justin Kamp / MIT.	On track
9/30/18	Draft journal article on evaluation of porosity profiles across the thicknesses of ceramic exhaust filter walls.	On track



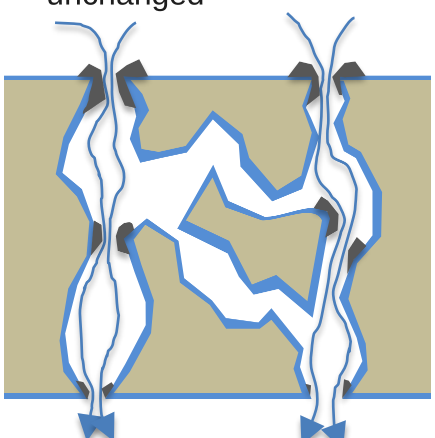
Approach

- Original scope and primary partner interest: fundamental understanding of relationship between substrate structure and filter performance
- Reviewers concerned that tools and approaches may not be applicable for extension to multifunctional (coated) and ash-aged filters

Extension of conceptual framework to coatings, ash

Light, uniform coating

- Examples: Some PGM-based oxidation catalysts
- Some pores constricted slightly
- Basic structure of pore network unchanged

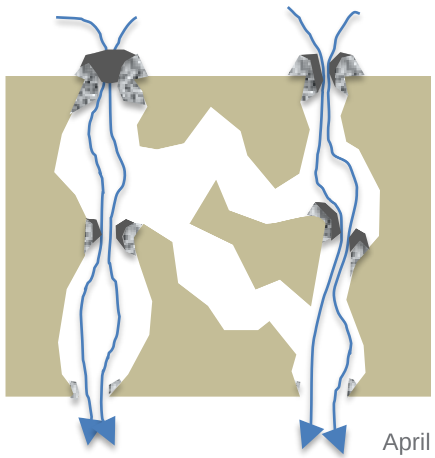


Heavy or non-uniform coating

- Examples: SCR-filter, TWC-GPF?
- Some pores filled or blocked
- Pathways through filter wall altered

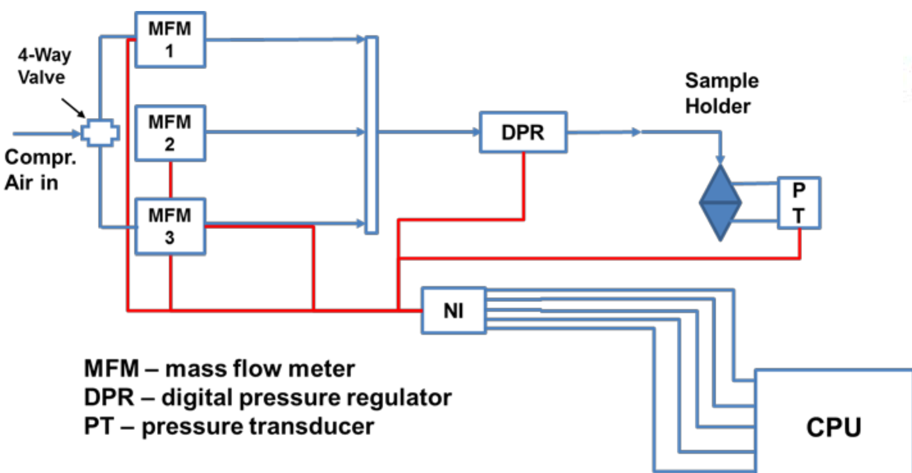
Ash

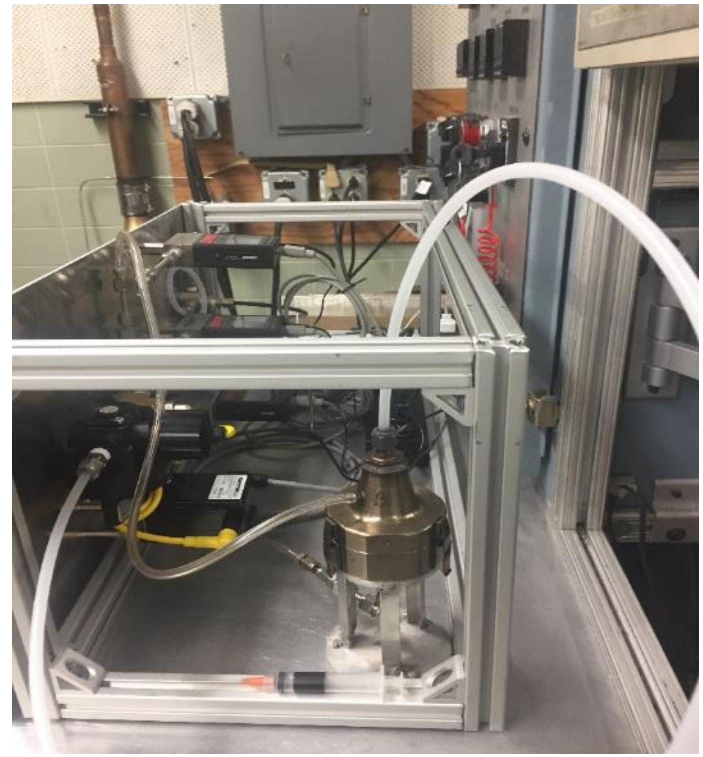
- Ash generated where soot is oxidized - more inside wall for GPF
- Ash particles consolidate, migrate, form hydrates
- Morphology varies, but ash typically porous, permeable

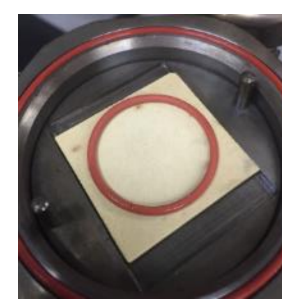


Accomplishments
Custom built CFP system for filter samples

- Capillary flow porometry setup built in-house, optimized for filter sample measurements
- Three mass flow meters to provide high accuracy over entire range of flowrates for measurements
  - 0-1 slpm, 0-10 slpm, and 0-100 slpm
- High-accuracy pressure transducer for specific pressure range of interest

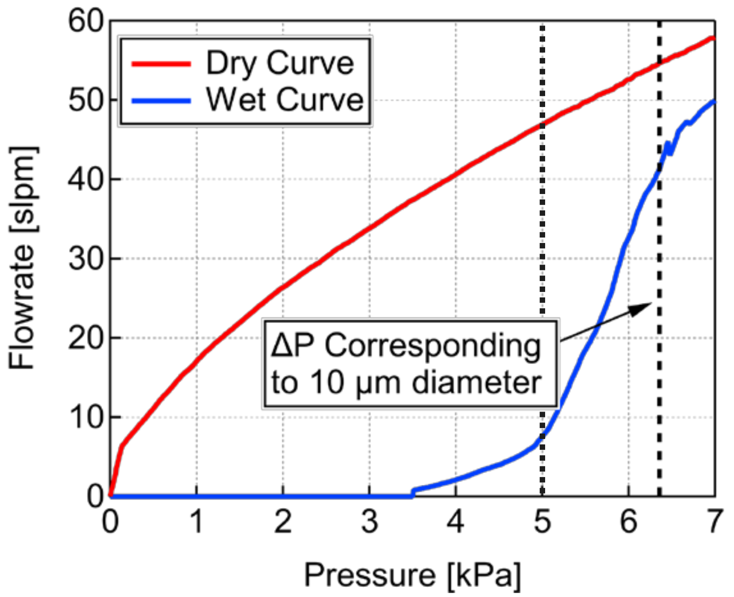






Accomplishments
CFP - validation

- Track etched polycarbonate samples used as validation case
  - Known pore size ~8-10 µm (narrow dist.)
  - Porosity = 5-20%
  - Thickness =16 µm
- In process of validating measurements
- Validated technique to be applied to all filter samples



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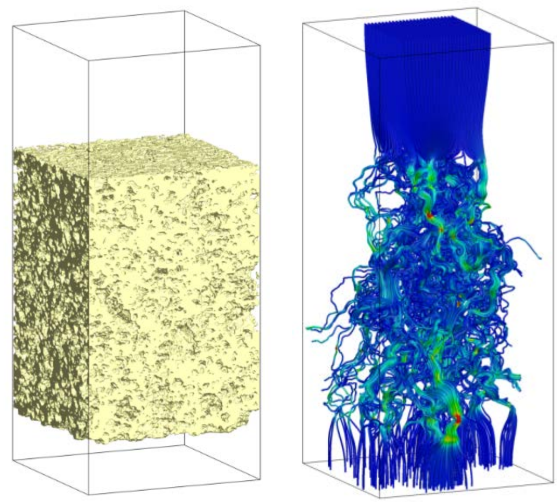
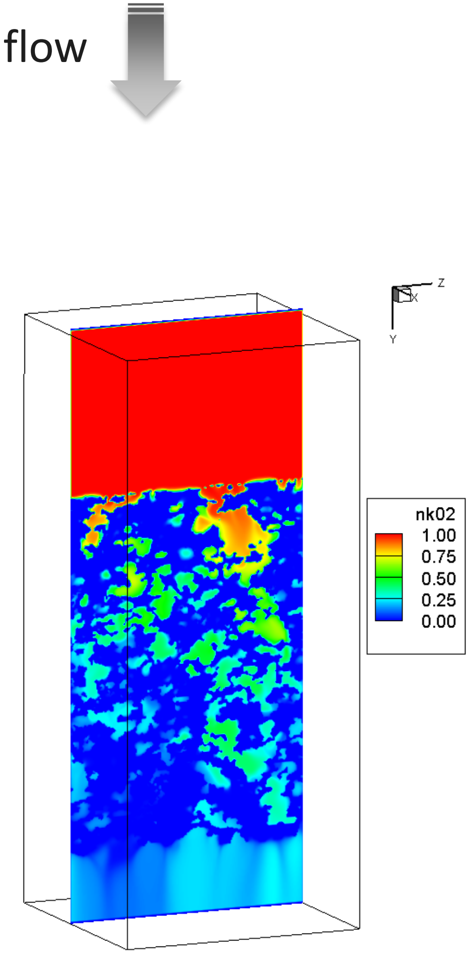
Accomplishments
Eulerian filter simulations using LB

- Started with LB flow solutions from last year
- Simulations run at 5.5 and 2.75 cm/s superficial velocity
- 50 nm particles
- Zero concentration boundary at solid surfaces within wall
- Results show which pores are efficiently removing particles and which are major pathways for penetration through the wall

Exhaust flow



C1



Normalized particle concentration

Number efficiency of a clean, fresh filter is an important limiting case for a production GPF – performance gets better with age

Accomplishments
Eulerian filter sims - seven substrates

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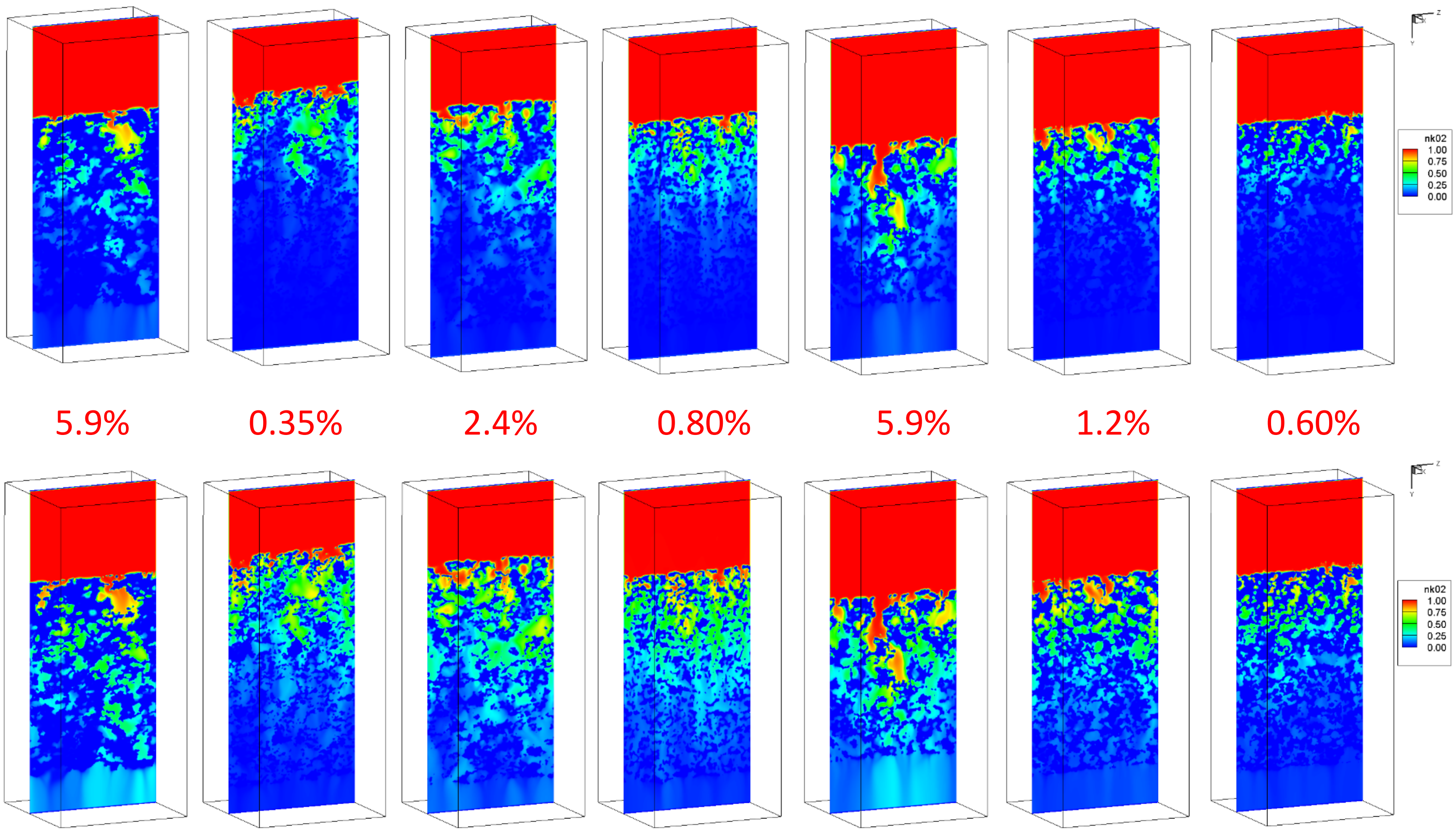

	C1	C2	C3	C4	C5	A1	A2
2.75 cm/s							
Penetration	5.9%	0.35%	2.4%	0.80%	5.9%	1.2%	0.60%
5.5 cm/s							
Penetration	16.9%	3.0%	9.0%	5.3%	16.6%	6.6%	4.8%

Accomplishments
Comparison of Eulerian sims to data

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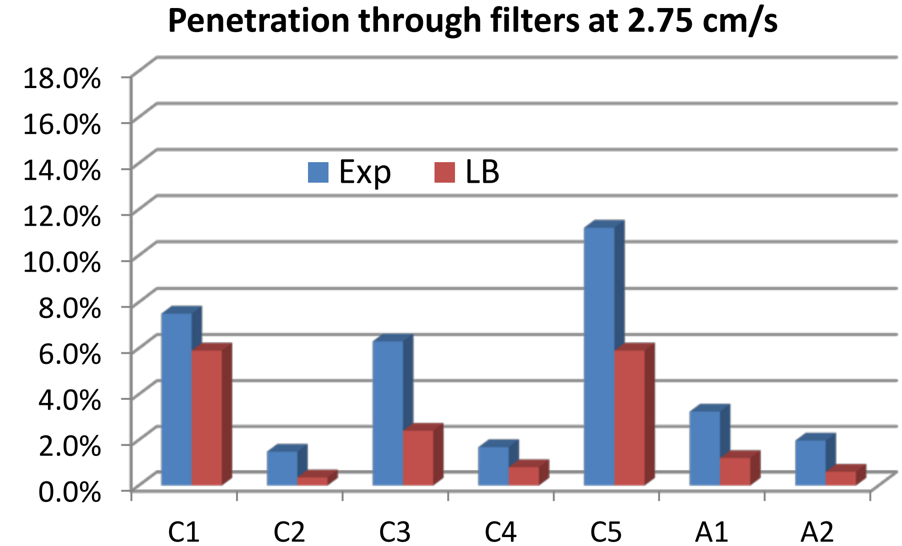
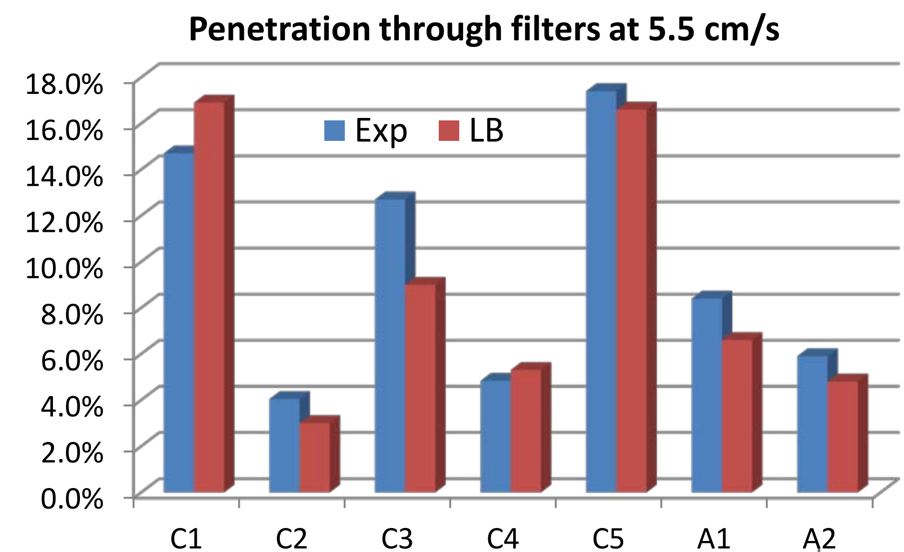
	Porosity	Median pore diameter (um)	Penetration: 2.75 cm/s Experiment	Penetration: 2.75 cm/s LB	Penetration: 5.5 cm/s Experiment	Penetration: 5.5 cm/s LB
C1	0.43	12	7.5%	5.90%	14.7%	16.9%
C2	0.65	17.9	1.5%	0.35%	4.0%	3.0%
C3	0.65	26.6	6.3%	2.40%	12.7%	9.0%
C4	0.67	17.6	1.7%	0.80%	4.8%	5.3%
C5	0.59	21.7	11.2%	5.90%	17.4%	16.6%
A1	0.53	17	3.2%	1.20%	8.4%	6.6%
A2	0.42	11.4	2.0%	0.60%	5.9%	4.8%





- Trends predicted at both flow rates
  - A2 efficiency correctly predicted much higher than C1
  - More penetration through substrates with larger pores, lower porosity
- Better quantitative match at 5.5 cm/s

Initial LB simulations fairly representative of experimental data *with no tuning*

Accomplishments
HMF and cylindrical pore models

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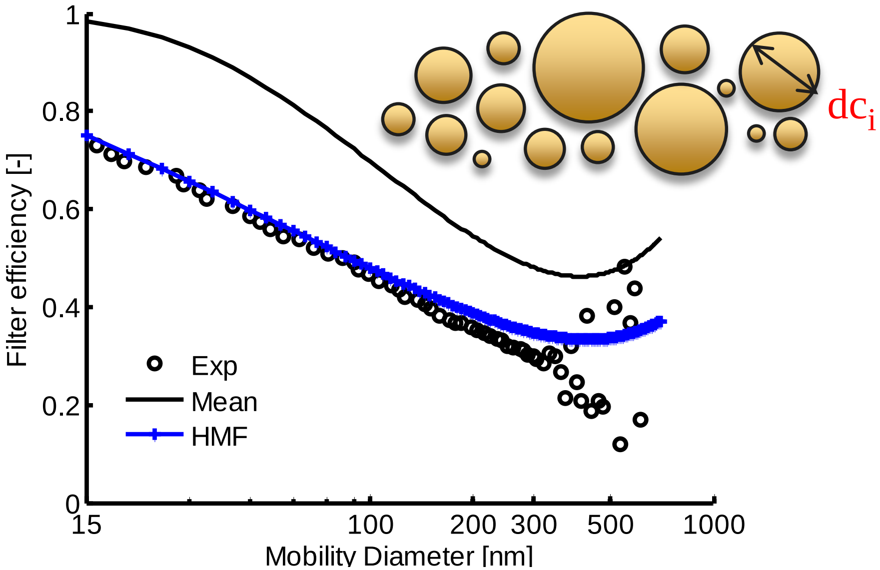
Heterogeneous Multiscale Filtration model

Jian Gong and Christopher Rutland

- Extension of traditional spherical unit collector model
- Distribution of collector sizes from mercury intrusion porosimetry (MIP)



Gong, J., et al., *Importance of filter's microstructure in dynamic filtration modeling of gasoline particulate filters (GPFs): Inhomogeneous porosity and pore size distribution.* Chemical Engineering Journal, 2018. 338: p. 15-26.

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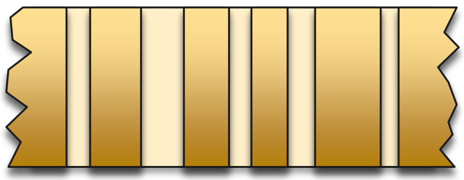
Cylindrical pore filtration model

Sandeep Viswanathan and David Rothamer

- Classical Karman representation of media by cylindrical pores
- Distribution of pore diameters from through-pores observed by CFP



50 nm particle penetration through wafer

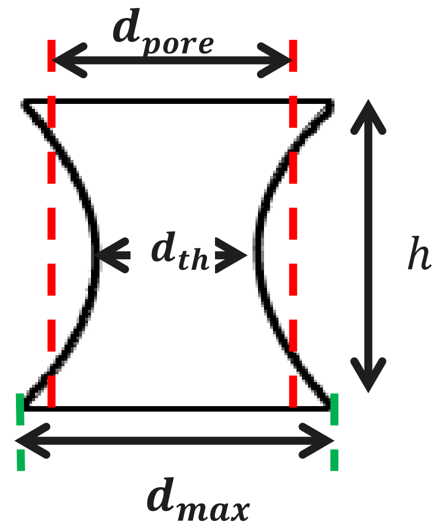
	C1		A2	
	2.75 cm/s	5.5 cm/s	2.75 cm/s	5.5 cm/s
Experiment (%)	7.6 ± 0.9	15 ± 3	2 ± 0.4	6 ± 1.5
Model (%)	7.6	14.6	2.2	5.6

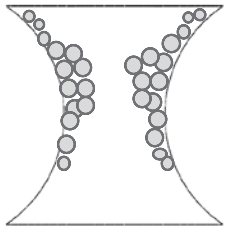
Viswanathan, S., et. al., *Experimental investigation of the effect of pore size distribution on nanoparticle capture efficiency within ceramic particulate filters*, 2018, (in preparation)

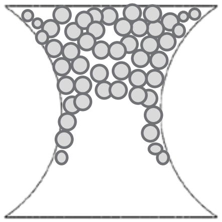
Accomplishments
Constricted tube collector model

- MIP → median pore diameter → d_{max}
- CFP → mean flow pore diameter → d_{th}
- h calculated based on specific surface area, assuming spherical grains

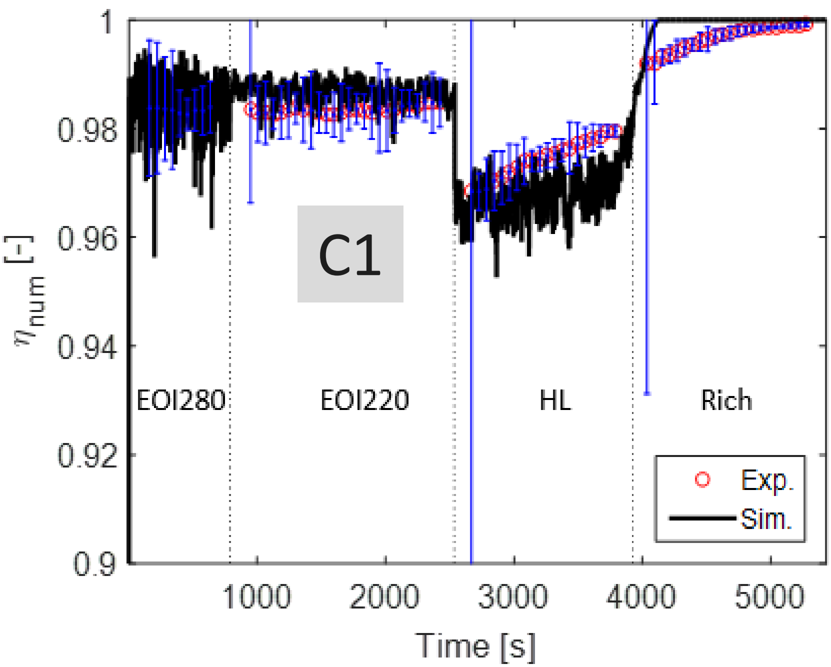
	Porosity (%)	MPD (µm)	Constriction (µm)	h [µm]	Perm [10^{-12} m^2]	Thickness [mm]
C1	43	12	5.88	23.8	0.68±0.1	1.05
C2	65	17.9	7.22	14.46	3.2±0.1	1.35
C3	65	26.6	11.21	21.48	7.3±0.7	1.40
C4	67	17.6	7.80	13	3.8±0.2	1.00
C5	59	21.8	10.35	22.72	4.3±0.4	0.90







Accomplishments
Constricted tube model – overall efficiency



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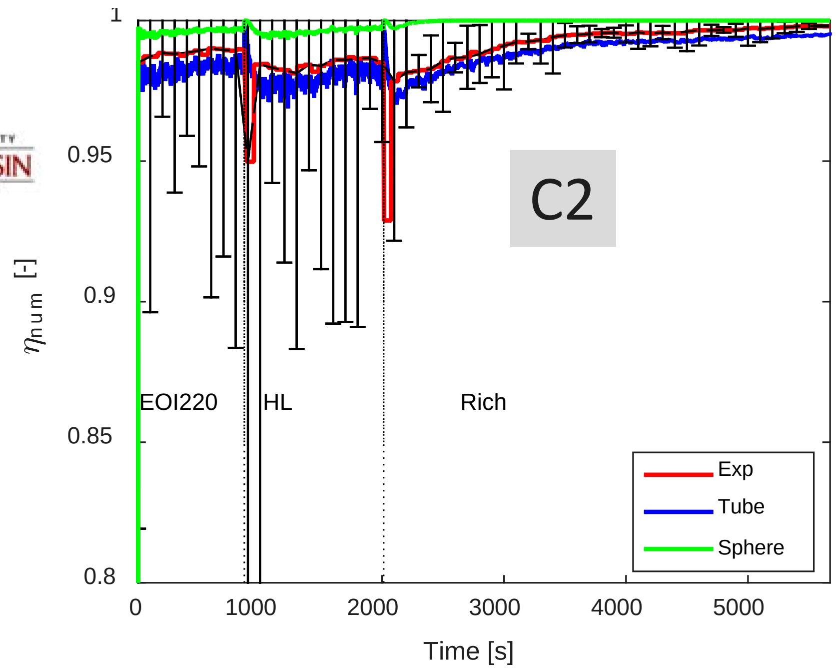

- Transient filtration experiments were carried out over a range of engine operating modes – different soot concentrations and size distributions
  - Progression from cleaner to dirtier modes led to more accumulation and progressively higher efficiencies
- The constricted tube model gives a reasonable match for overall number efficiency as soot accumulates within the wall – better than the traditional spherical collector model

Constricted tube model – size resolved efficiency and backpressure

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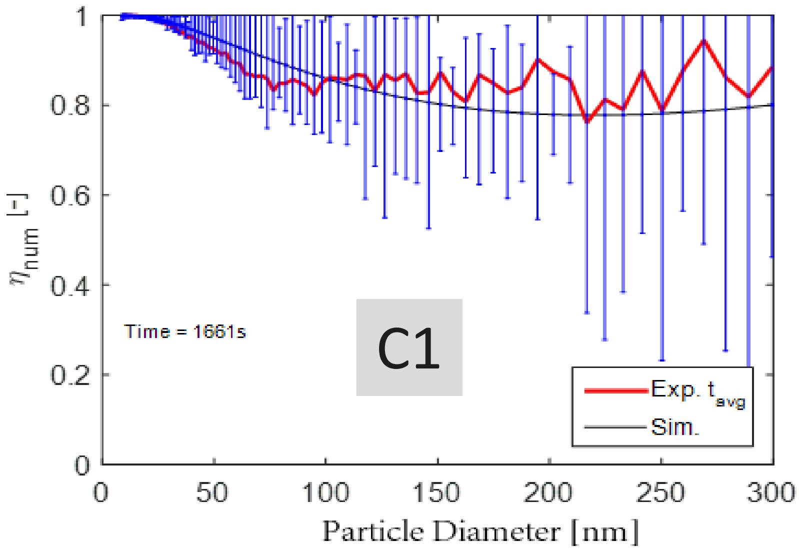
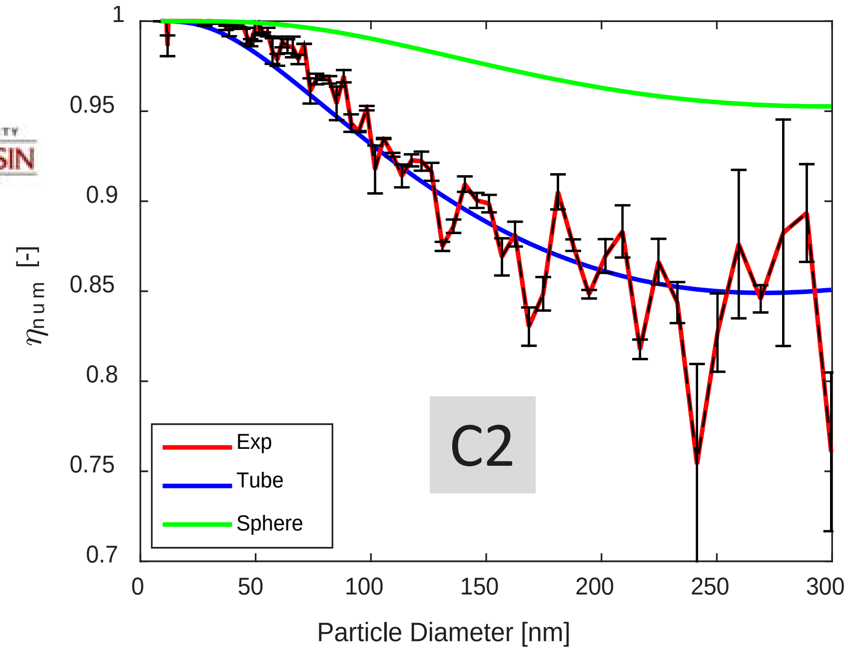
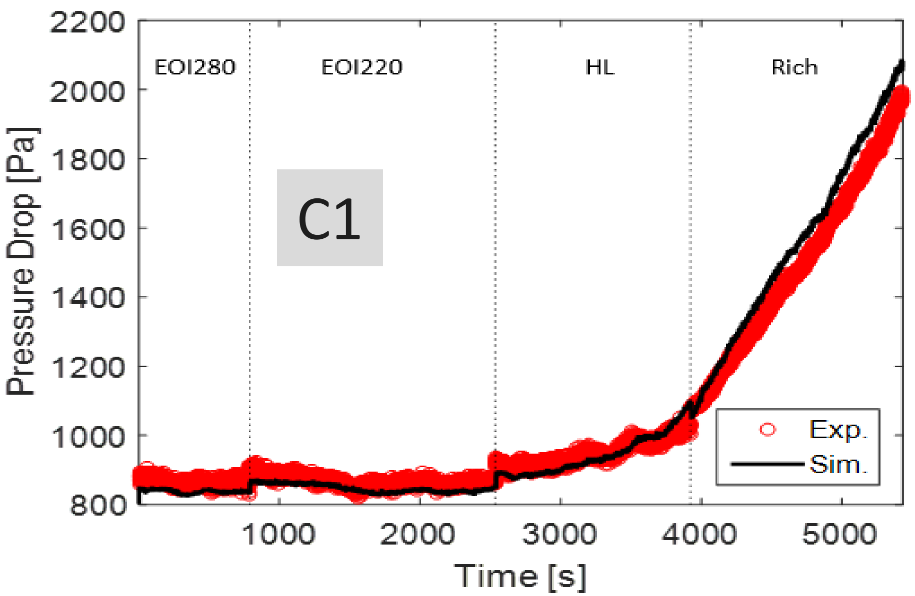


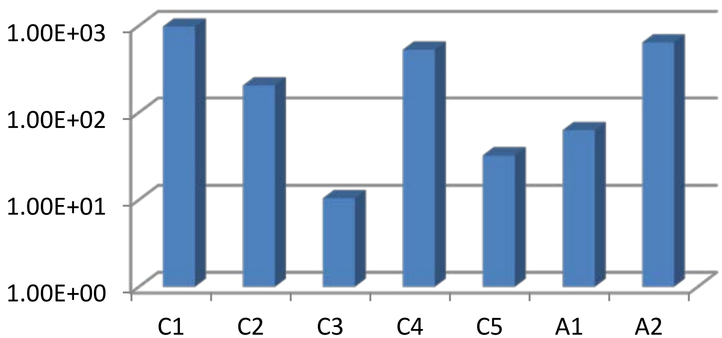




- The constricted tube model also matches size-resolved filtration efficiency and backpressure as a function of loading

Accomplishments
Modified spherical collector model

Ratio of experiment to predicted penetration
5.5 cm/s – Classical diffusion efficiency

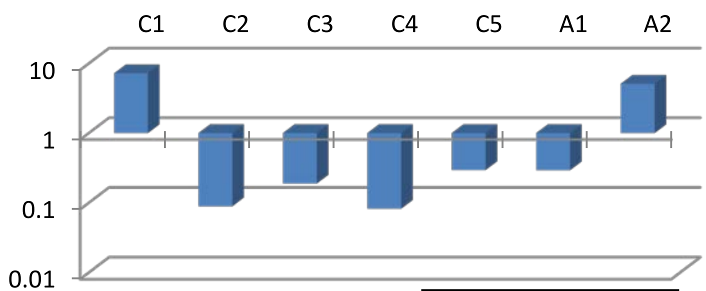


$$\eta_D = 4.0 A_s \left(\frac{1}{Pe} \right)^{2/3}$$

$$A_s = \frac{2(1 - p^5)}{2 - 3p + 3p^5 - 2p^6}$$

$$p = (1 - \varepsilon)^{1/3}$$

Ratio of experiment to predicted penetration
5.5 cm/s – Long and Hilpert diffusion efficiency



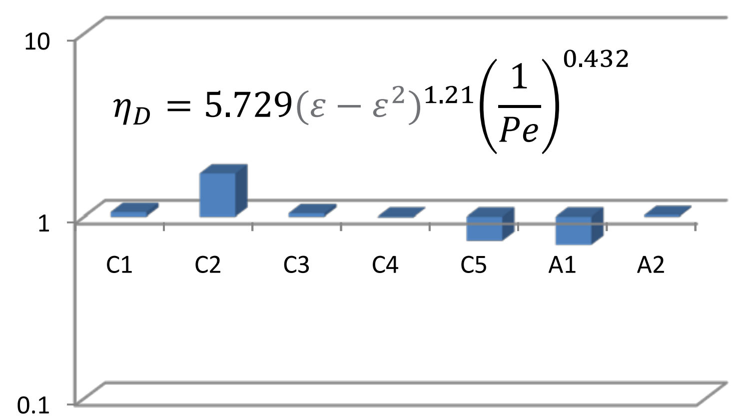
$$\eta_D = 15.46 \frac{(1 - \varepsilon)^2}{\varepsilon^3} \left(\frac{1}{Pe} \right)^{0.65} R^{0.19}$$

$$R = \frac{d_{particle}}{d_{collector}}$$

Long, W. and M. Hilpert, ENVIRONMENTAL SCIENCE & TECHNOLOGY, 2009. 43(12): p. 4419-4424.

- The traditional spherical unit collector model over-predicts filter efficiency by orders of magnitude for 50 nm particles
- A modified expression from the literature does a better job
  - Optimized for loose pack of spheres using LB simulations
- We are developing new expressions – optimized for ceramic exhaust filters

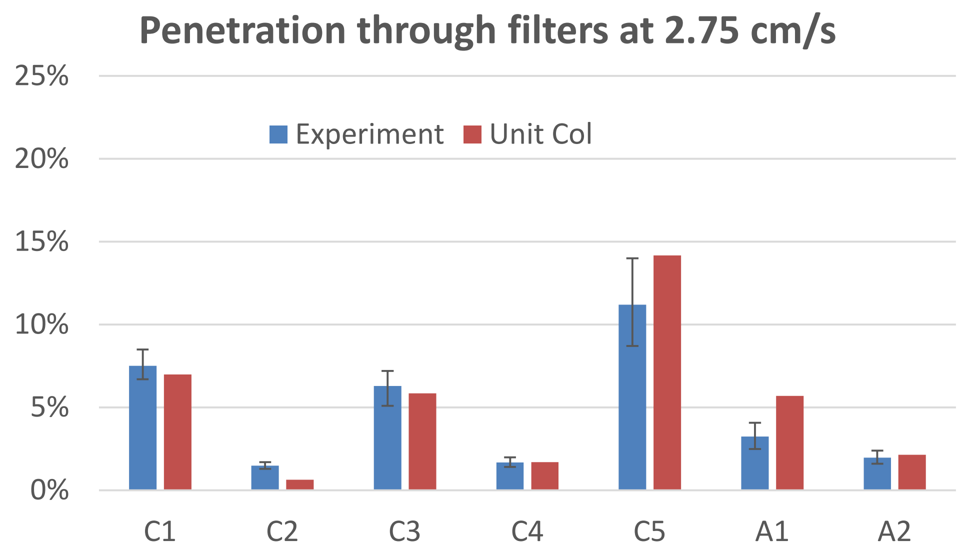
Ratio of experiment to predicted penetration
5.5 cm/s – New diffusion efficiency expression

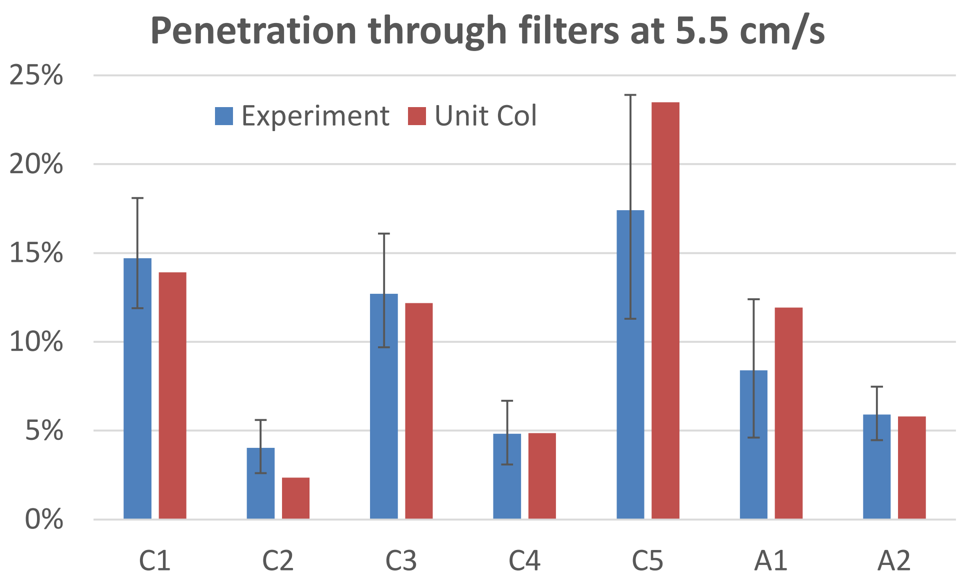


$$Pe = \frac{u_s \cdot d_{collector}}{D_{particle}}$$

ε = filter porosity

Accomplishments
Modified spherical collector model





- A number of new diffusion efficiency expressions are being evaluated
- Expressions can be optimized using either the experimental filtration data for the wide range of substrates studied or lattice-Boltzmann simulation results
- The model shown here also uses a new method for calculating the collector size, which takes into account the shape of the pore size distribution

Accomplishments
Evaluating effects of ash and coatings

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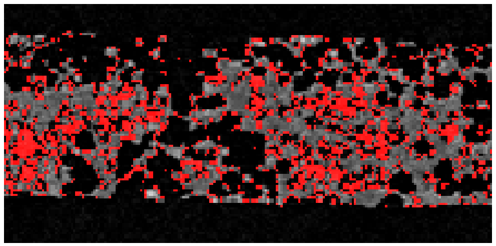
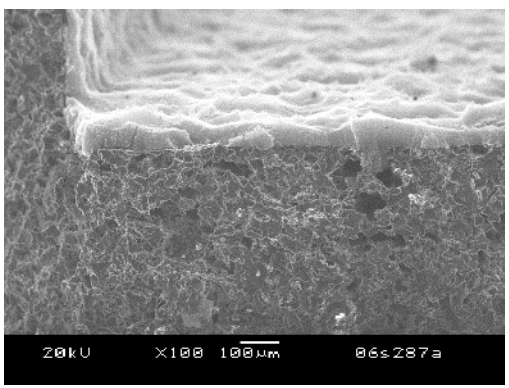
- Effects on filtration will depend on substrate and nature of ash or catalyst loading
- Methods for studying the effect of structure on flow paths will still be applicable
- Cooperative studies with Justin Kamp are now looking at ash and coating effects on flow and permeability

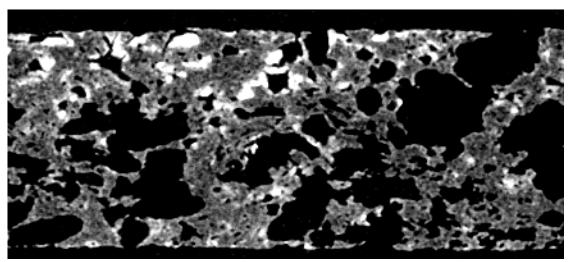


SCR on cordierite
Some pores blocked

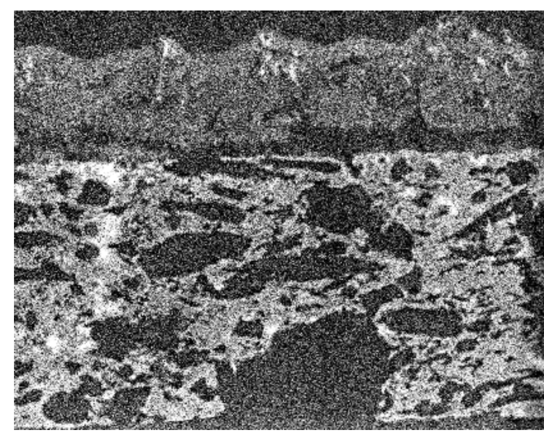
Advanced oxidation catalyst on mullite
Thin coating does not fill pores



Oxidation catalyst on cordierite
Some surface pores covered

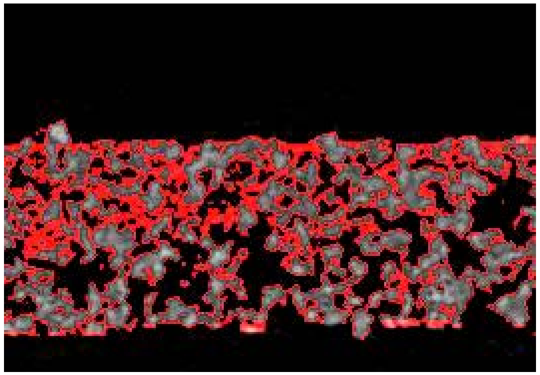


Oxidation catalyst on cordierite (MIT)
Catalyst brighter than substrate
Fills some pores near one wall surface



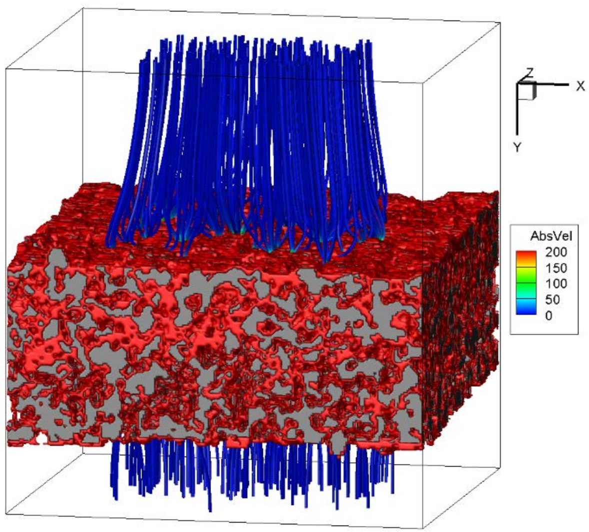
Diesel ash on cordierite (MIT)

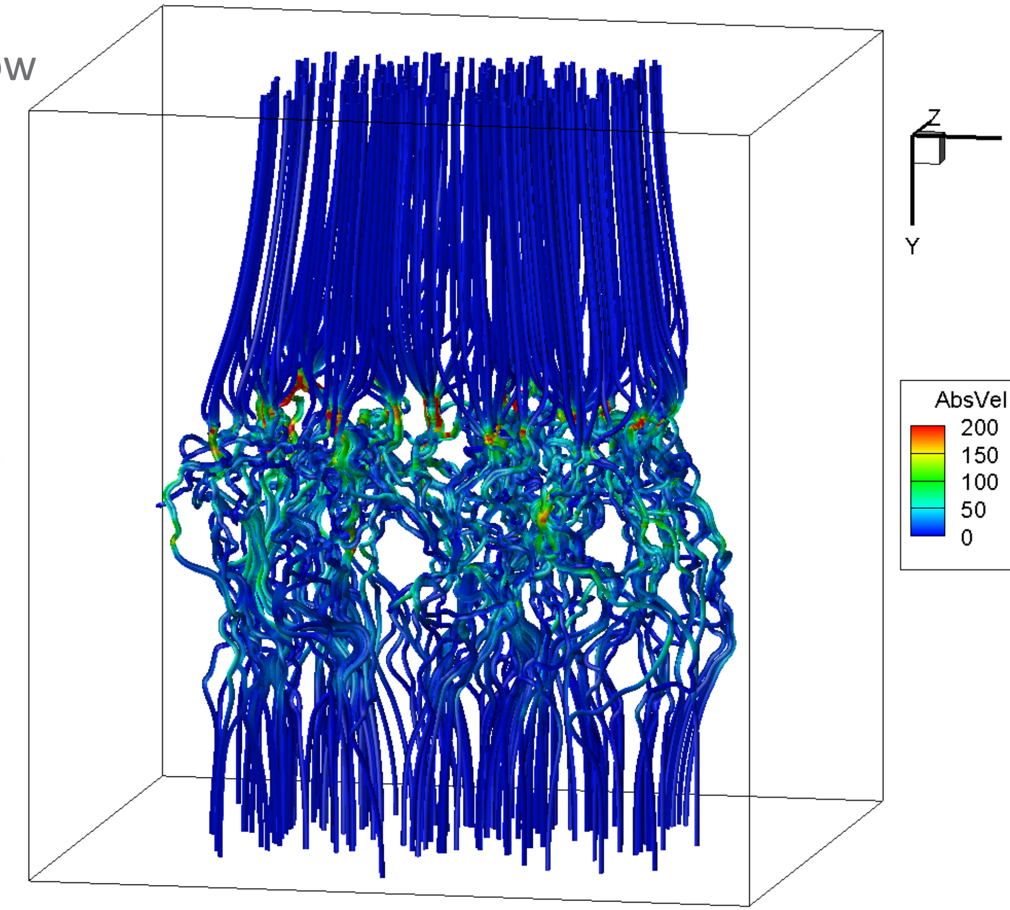
Accomplishments
Evaluating effects of catalyst coatings



Exhaust flow

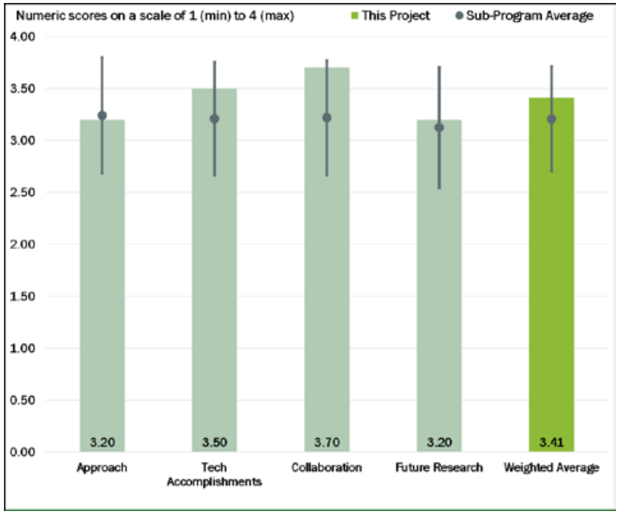
Catalyzed filter wall





- SCR on SiC filter exhibits gradient in catalyst loading across wall
- LB simulations show fewer flow paths, higher velocities where catalyst loading is heaviest

Responses to 2017 reviewer comments



- **"This is a good fundamental research project."**
- **"… the approach of characterizing the particles and filter porosity and then correlating this to filter performance is generally excellent."**
- **"… great progress in converting the spherical unit collector model to the constricted tube model…"**
- **"… technical work on filter characterization is nice and detailed."**
- **"… team is certainly pushing the analytical technology to new horizons with this world class work."**
- **"… it is conceivable that all engines will have filters and filters can have a big impact on engine efficiency. This team is developing fundamental knowledge to help."**

- "… team needs to be cognizant that fresh filters are applicable only for the first thousands of miles, and then ash begins impacting efficiency and back pressure."

  This is an important point, and while ash aging was not in our original scope, we are trying to develop tools that can be extended to aged devices.

- "The reviewer recommended that the study include both uncatalyzed and catalyzed filters."

  Some characterization of catalyzed filters has been completed and experiments are planned.

- "Emissions data on a SIDI engine were mentioned yet no connection was made to improved filtration ability or if a filter is still needed."

  Although we have focused on SIDI, from the beginning this project was intended to be fuel-agnostic. The goal has been to develop fundamental characterization and modeling tools that can be broadly applicable as engines, aftertreatment technology, and regulations develop. Our hope is that improved understanding and models well help OEMs and device manufacturers better negotiate the many trade-offs inherent in aftertreatment system design.

Collaboration and coordination

- Major Partners
  - General Motors Company (Industry): Provide funding (supporting full-time graduate student working on improved models), hardware, expertise, and operational guidance for work at the University of Wisconsin, Madison. Advise on project direction and priorities.
  - Engine Research Center at University of Wisconsin, Madison (Academic): Operate test engine and EFA system - including shakedown tests, independent experiments, and cooperative experiments. Assist in analysis and publication of data. Develop improved device-scale modeling techniques.
  - Massachusetts Institute of Technology (Academic): Perform X-Ray CT scans, including ultra-high (sub-micron) resolution. Provide access to datasets for catalyzed and ash-aged parts. Capabilities also include advanced artifact reduction and segmentation tools.

- Analysis subcontracts
  - Micromeritics
  - Particle Tech Labs
  - Micro Photonics

- Filter suppliers
  - Corning Incorporated
  - Ibidin
  - NGK
  - Sumitomo

Remaining challenges and barriers

- Filtration data for continuous regeneration conditions at high temperature and low PM loading (representative of close-coupled GPF) is needed to validate relevant models.
- More general models are needed, which will allow prediction of filter performance as a function of well-defined structural properties over a wide range of engine operating conditions, especially for the removal of the very small particles at low PM loadings expected for gasoline applications.
- Better fundamental understanding is needed of the way catalyst coatings and ash alter the behavior of various filter substrates, especially under conditions representative of emerging applications.

Sixth generation Chevrolet Camaro
2016 Geneva Auto Show
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Planned future work

- Wrapping up this project
  - Complete capillary flow porometry (CFP) studies of at least 7 substrates with custom-built system optimized for exhaust filters
  - Complete high-temperature EFA filtration experiments
    - Evaluate effects of volatile organics
    - Evaluate filter performance during continuous regeneration (analogous to close-coupled unit)
  - Complete development of constricted tube collector model
  - Complete cooperative study with Justin Kamp of coated and/or ash aged filter permeability from micro-scale flow simulations
  - Extend new diffusion expressions for spherical unit collector model
    - Expand range of physical properties using hypothetical digital materials simulated with LB
    - Validate expression over a range of particle sizes
  - Publish findings
- Possible follow-on projects
  - Extension of filter performance experiments and model development to multi-functional devices and/or filters ash-aged under conditions representative of advanced engines
  - Focus on compression ignition gasoline particulate characterization and aftertreatment

Any proposed future work is subject to change based on funding levels

Summary

- Custom-built capillary flow porometry (CFP) system
- Re-built test engine and EFA cart in preparation for high-temp filter experiments
- Evaluated modeling, analysis tools for future extension to coated, ash-aged filters
- Multiple modeling studies:

Models Used / Developed	Substrates Examined	Demonstrated Predictions	Notes	Dissemination
Lattice Boltzmann	C1, C2, C3, C4, C5, A1, A2	Clean removal efficiency for small particles, 2 flow rates	First principles – no tuning, use 3D X-Ray scans	CLEERS workshop, journal article in prep
Heterogeneous Multiscale Filtration	C1, others	Evolving size-resolved efficiency and pressure drop during filtration	Pore size and distribution from MIP, porosity distribution from X-ray CT	Dissertation, presentations, peer reviewed publications
Cylindrical tube	C1, A2	Clean removal efficiency for small particles, 2 flow rates	Pore throat size distribution from CFP	Dissertation, SAE oral, journal article in prep
Constricted tube	C1, C2, C3, C4, C5	Evolving size-resolved efficiency and pressure drop during filtration	Pore size distribution, shape from MIP, CFP	SAE paper, dissertation in prep
New spherical collector	C1, C2, C3, C4, C5, A1, A2	Clean removal efficiency for small particles, 2 flow rates	Pore size distribution from MIP	

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Technical Backup Slides

Exhaust Filtration Analysis (EFA) experiments

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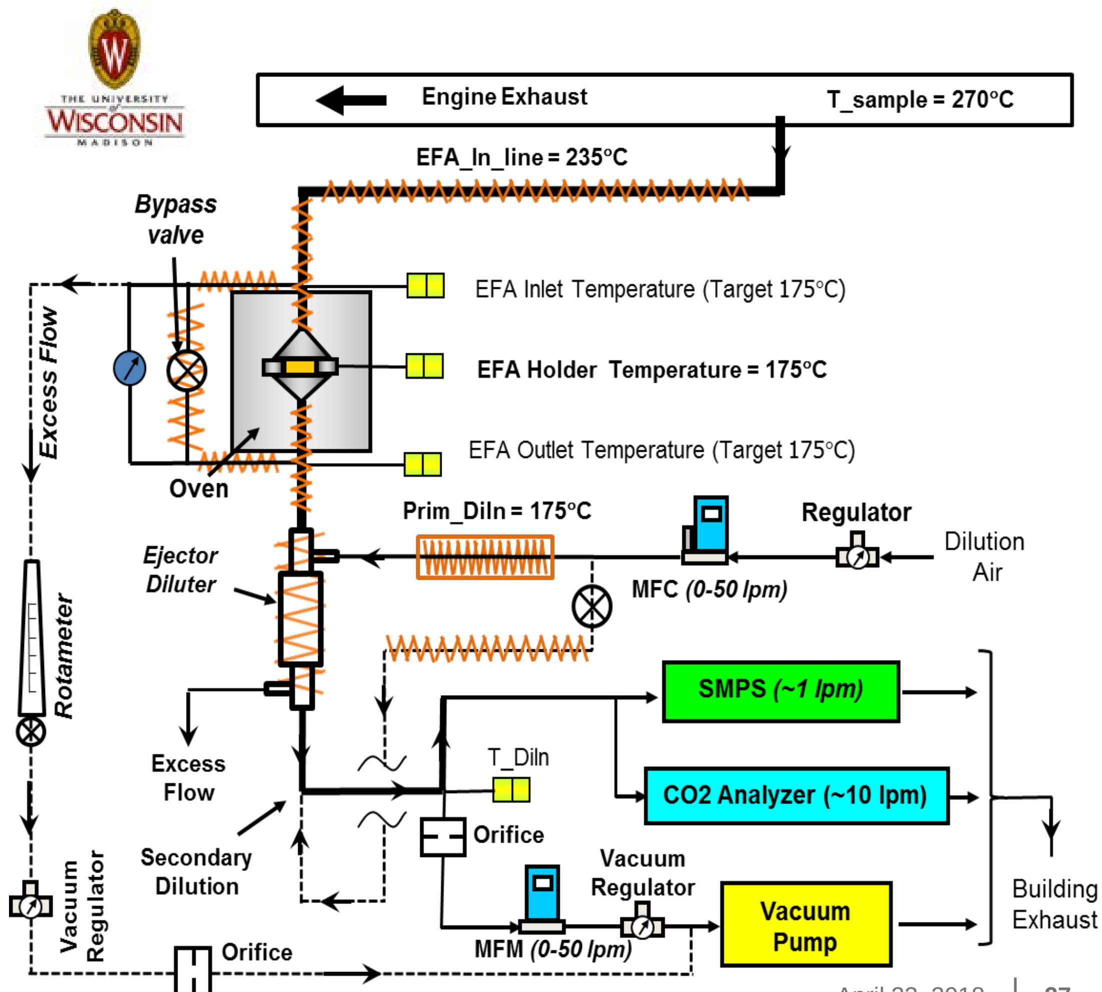
GM / UW-Madison Collaborative Research Laboratory

- Filtration experiments conducted with flat wafer samples and exhaust from single cylinder test engine
- Particulates measured with Scanning Mobility Particle Sizer (SMPS) and Engine Exhaust Particle Sizer (EEPS)

See SAE-2014-01-1558



Filter properties summary

(for Reference only)

Batch	Porosity (%)	σ	MPD (μm)	SD [μm]	W	Perm. [10^{-12} m^2]	Thickness [mm]
C1	43	0.31	12	4.45	0.53	0.68±0.1	1.05
C1-C	43	0.27	13.4	2.47	0.44	0.50±0.008	1.05
C2	65	0.13	17.9	2.55	0.55	3.2±0.1	1.35
C3	65	0.16	26.6	4.89	0.52	7.3±0.7	1.40
C4	67	0.11	17.6	2.07	0.36	3.8±0.2	1.00
C5	59	0.22	21.8	5.51	0.49	4.3±0.4	0.90
C6	65	0.26	14.6	3.90	0.46	1.27±0.09	0.31
C7	64	0.14	16.7	2.38	0.17	1.98±0.06	1.01
C8	64	0.21	14.3	3.21	0.34	1.81±0.03	1.01
C9	62	0.31	13.6	4.52	0.47	TBD	0.26
A1	53	0.15	17	2.81	0.39	3.0±0.15	1.00
A2	43	0.07	11.4	0.82	0.24	0.76±0.1	1.05

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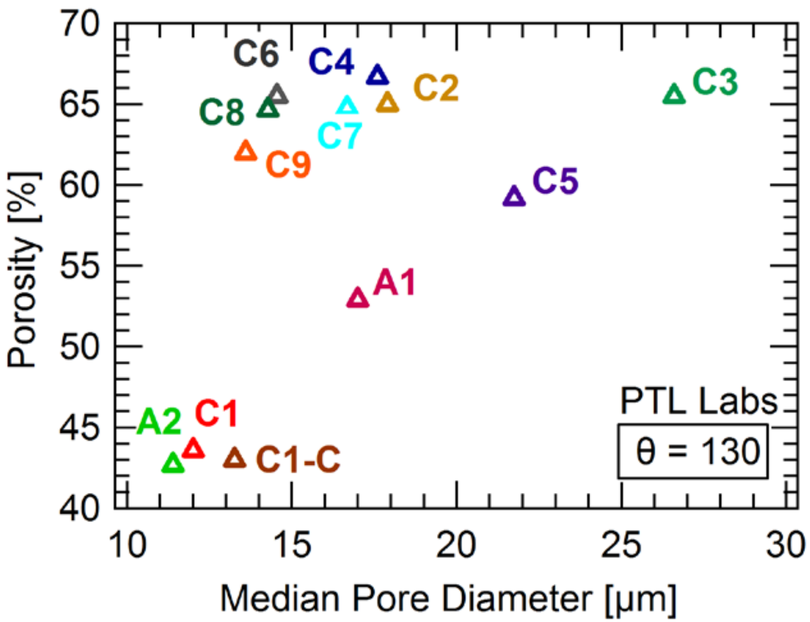

Porosity vs. Median Pore Diameter

Cordierite (C) and Aluminum Titanate (A) samples tested. **Newly tested filter samples**.

Filter Characterization

Capillary Flow Porometry (CFP)

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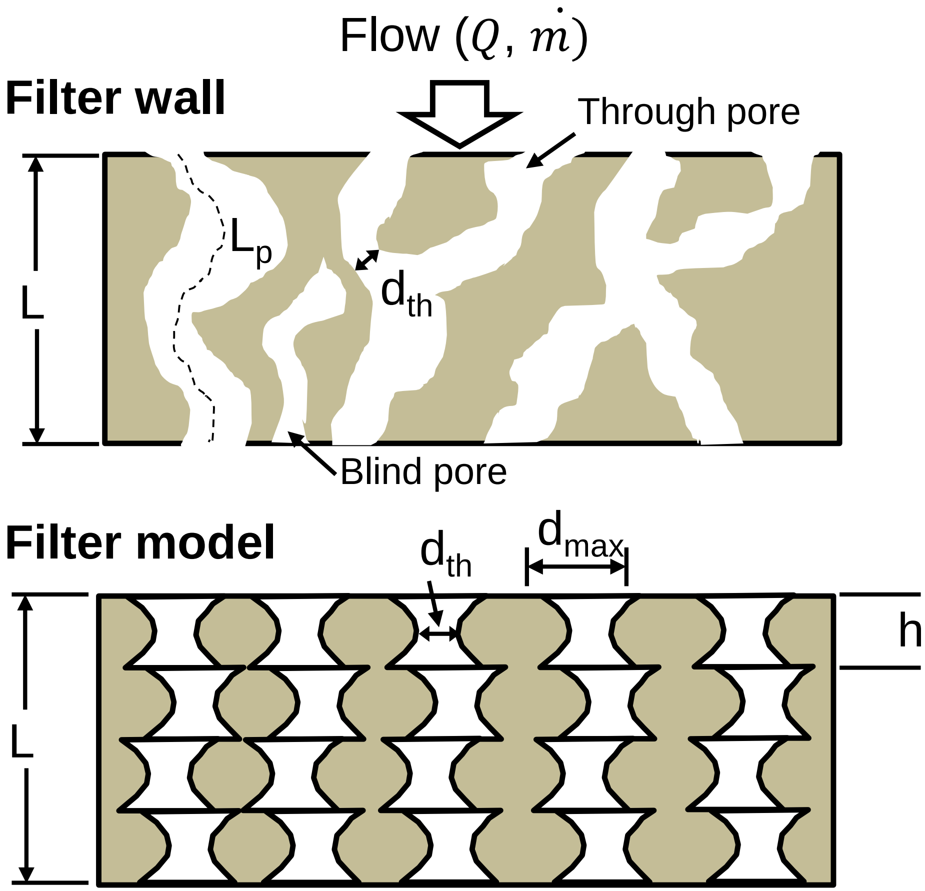
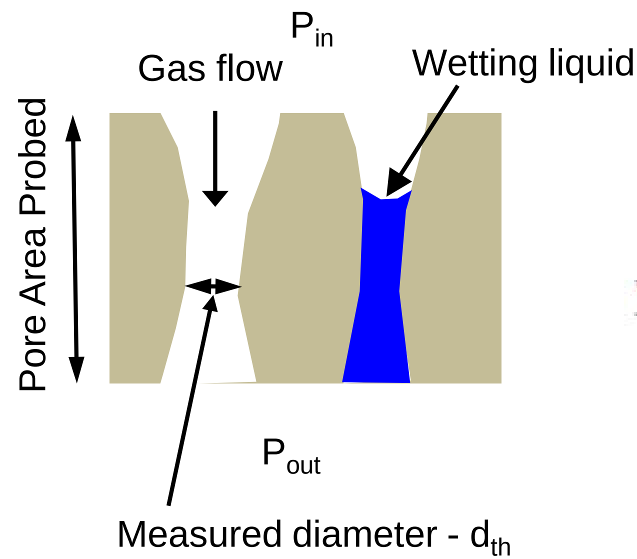


Schematics illustrating (top) realistic filter morphology and (bottom) constricted tube model morphology

Capillary Flow Porometry



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- CFP directly provides information on throat diameter needed for pore filtration model
- CFP can provide data on the distribution of throat sizes in samples

Efforts to quantify participating flow volume using LB simulations

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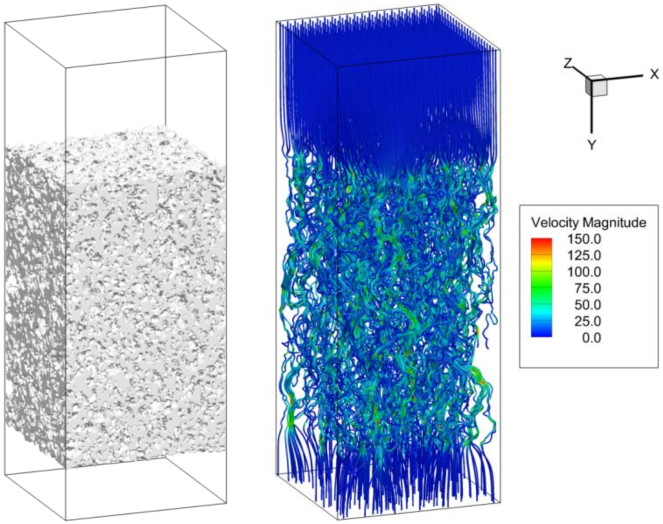
- Substrates with worse pore connectivity direct more of the flow to relatively fewer major pathways through the filter walls, effectively bypassing some of the pore volume
- One idea for quantifying the difference:
  - Look at the absolute velocity magnitude at each pore voxel within the filter wall
  - Count the voxels with velocity below some threshold – here I used percentages of the average interstitial velocity for each substrate
  - Divide the count by total pore voxels to find the fraction of pore volume with velocity below the threshold



$$v_{avg} = \frac{v_{face}}{porosity} \qquad v_{face} = 2.75\,\frac{cm}{s}$$

			Percent of void volume at less than a given percent of v_{avg}			
	v_{max} (cm/s)	v_{avg} (cm/s)	1%	2%	5%	10%
C1	423.9	6.405	11.29%	14.61%	20.58%	26.29%
C2	73.45	4.240	1.50%	2.40%	4.81%	8.62%
C3	77.45	4.269	2.30%	3.67%	7.07%	11.89%
C4	50.75	4.140	0.63%	1.23%	3.08%	6.34%
C5	130.4	4.688	2.19%	3.60%	7.10%	12.05%
A1	116.7	5.224	2.04%	3.40%	6.81%	11.50%
A2	234.5	6.493	3.24%	4.88%	8.63%	13.41%

Eulerian filter simulation concentration and flux profiles

- “Flux” is the average advective particle flux in the y direction
- Within the wall the flux profile is fit very well by exponential decay